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Frustrated magnetic moments in small metal clusters

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Abstract The magnetic properties of small antiferromagnetic clusters with nearest neighbour interaction containing atoms upto 561 have been studied The magnetic moments of small antiferromagnetic clusters have been investigated as a function of size, temperature and magnetic field. The calculations for icosahedral and cuboctahedral clusters have been carried out self-consistently by assuming that the clusters undergo super paramagnetic relaxation. The effective magnetic moment (μ_{eff}) per atom of icosahedral clusters increases with magnetic field but decreases with increasing temperature It is observed that μ_{eff} of icosahedral clusters reaches saturation value with magnetic field while that of cuboctahedral clusters remains unchanged with the magnetic field for N = 13 and 55 atoms in a cluster. It has been analyzed in both icosahedral and cuboctahedral clusters that μ_{eff} increases linearly with cluster size before saturating at larger size.

Keywords Antiferromagnetic clusters, super paramagnetic relaxation, effective magnetic moment, mean field theory, magnetic field, ferromagnetic fine particles

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One of the most fundamental problems in cluster research is to understand how the Physical and Chemical properties of clusters get changed owing to the variation of temperature, size and magnetic field [1-4]. The change in atomic environment, affects the itinerancy, electronic correlations and spin-orbit coupling, leading to a close inter-dependence of magnetism and cluster structure for the increasing cluster size. The icosahedral and cuboctahedral clusters are expected to be the most stable lattice structures at low temperatures, so that the magnetic moments of antiferromagnetic clusters can be explained in detail [5]. The magnetic properties of small antiferromagnetic clusters such as the changes of the temperature and the magnetic moments, which are changed due to the cluster size, are the major problems for many investigations [6].

Reddy and Khanna [5] have proposed the Heisenberg model to investigate the magnetic behaviour of clusters with antiferromagnetic interactions. The frustrated paramagnetism was observed in the icosahedral clusters containing atoms between 13 and 561 and it has been proved theoretically that the magnetization of antiferromagnetic clusters can be determined using Ising spins [5]. In this paper, the chosen problem addresses the existence of antiferromagnetism in superparamagnetic model and new properties of clusters that are not observed from the bulk magnetic properties. The main reason to investigate the effective magnetic moment of icosahedral and cuboctahedral clusters is that they exist in almost all the known metal clusters. In the present work, it has been tried to explain the magnetic field and the exchange interaction to support the validity of the effective magentic moments.

The localized spins containing two possible values $S_1 = \pm 1$, located at each atomic site have been addressed by the site index S_i. The Hamiltonian of the system is given by [2,6,5]

$$H = J\sum_{(i,j)}^{..} S_i . S_j - B\sum_{i=1} S_i \,. \tag{1}$$

Here, the summation in the first term is carried over to the nearest neighbour with external magnetic field B and gives the superparamagnetic relaxation for the small antiferromagnetic clusters. The external magnetic field H_j is taken with $g\mu_B$. J is the exchange interaction or coupling constant having the value less than zero for antiferromagnetic coupling. Owing to the absence of data, J has been treated as an adjustable parameter. On the other hand, only qualitative experimental information is available on J with the help of which it has been adjusted [2]. It is assumed that the antiferromagnetic clusters undergo superparamagnetic relaxation. The μ_{eff} per atom in the direction of the field H_j or an assembly of clusters at a temperature T is calculated.

The internal magnetic field at each spin site (j) is given by the mean field equation [7]

$$H_j = \sum J(j-j')\langle Sz_j \rangle, \tag{2}$$

where $J(n)$ is the spin-spin interaction with its n^{th} neighbour. $Sz_{j'}$ is the thermally averaged mean value of the spin component for the magnetic ions in the j^{th} site.

The single domain clusters having N atoms will have N_μ moment, where μ is the atomic magnetic moment. For small magnetic anisotropy, one can expect the cluster moment (N_μ) to exhibit a Boltzmann distribution of orientations with respect to H_j at thermal equilibrium. The effective magnetic moment described by the Langevin function of icosahedral and cuboctahedral clusters is given by

$$\mu_{eff} = \mu B_j(x), \quad x = \frac{N\mu H_j}{kT} \tag{3}$$

The surface effects are taken into account by taking N and N_{eff} in eq. (3) and is given by

$$N_{eff} = \frac{N}{2}\left[1+\left(1 - \frac{Ns}{N}\right)\right] \tag{4}$$

Here, Ns are the number of atoms residing on the surface. The effect of surfaces is not geometric only. However, eq. (4) has been used in the absence of a rigorous theory, which includes the surface effect through a geometry-dependent empirical relation. The self-consistent calculations have been carried out using eq. (3). The thermal behaviour of the antiferromagnetic clusters has been analyzed within the classical Heisenberg model using Monte-Carlo simulations [8, 9].

In Figure 1, we have presented the computed values of μ_{eff} of icosahedral clusters in terms of μ_B for T=0.2.K. The μ_{eff}'s have been calculated for N = 13,55,147,309 and 561 atoms in a cluster with respect to the field. It is interesting to note that the μ_{eff} increases linearly at low fields. As the field increases further, the μ_{eff} reaches the saturation value. The saturated μ_{eff} values for N = 13,55,147,309, and 561 are 0.2, 0.2848. 0.2673, 0.1678 and 0.1567 μ_B , respectively. The decreasing μ_{eff} exists from the nearest neighbour interaction with external magnetic field; which tends to align all spins in the same direction. The decreasing trend in the μ_{eff} values has been compared with values calculated by Reddy and Khanna [5]. They have reached the saturated μ_{eff} values at 0.23, 0.24, 0.22, 0.105 and 0.09 μ_B respectively for N = 13, 55, 147, 309 and 561.

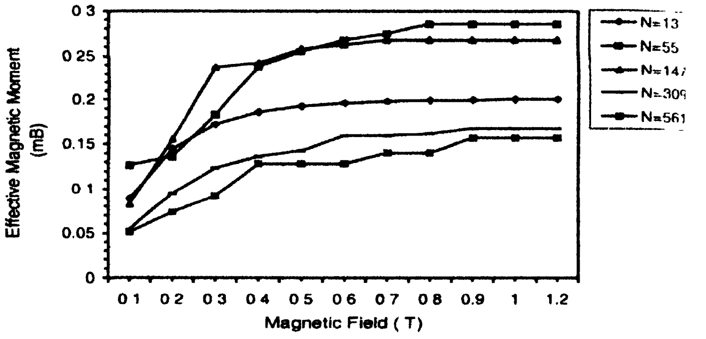

Figure 1. Size and structural dependence of the magnetic properties of icosahedral clusters for different applied fields with T = 0.2 K.

The decreasing μ_{eff} is used to identify the antiferromagnetic interaction between the nearest neighbour spins. This has been observed using Monte Carlo study with the lowest energy configuration of the icosahedral clusters [5]. The antiferromagnetic interaction between the nearest neighbour spins is dominated at very low temperatures and they aligned in the opposite directions [6]. In small antiferromagnetic clusters, it happens that the two spins (±) pointing in the two directions are not equal. Therefore, this leads to the nonvanishing magnetic moment both at low temperatures and in low external magnetic fields. The paramagnetism at high temperatures has been observed with H = 0.2 T and is plotted in Figure 2.

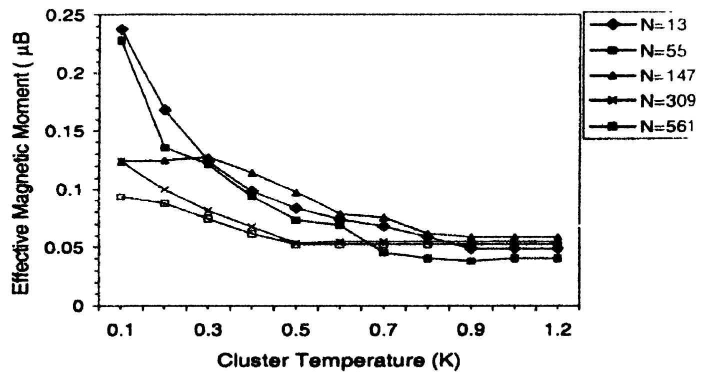

Figure 2. Size and structural dependence of the magnetic properties of icosahedral clusters for different temperatures with H = 0.2 T.

In Figure 3, we have given the computed values of cuboctahedral clusters having atoms between 13 and 561. μ_{eff}'s have been calculated for various magnetic fields with T = 0.2 K. If we increase the field B, the μ_{eff} of 0.2738 μ_B remains unchanged for N = 13 atoms. It shows that there is no presence of free spins in a cluster for this cluster size. For all other cases too, μ_{eff} is almost constant with N = 55,147, and 561 atoms. It has been observed that the exchange interaction J is lower for smaller antiferromagnetic clusters than the ferromagnetic clusters. It is generally believed that J is unknown and it is low for antiferromagnetism [6]. Again, μ_{eff} decreases as the cluster size increases with field for fixed temperature value. The decreasing μ_{eff} with increasing temperature is given in Figure 4.

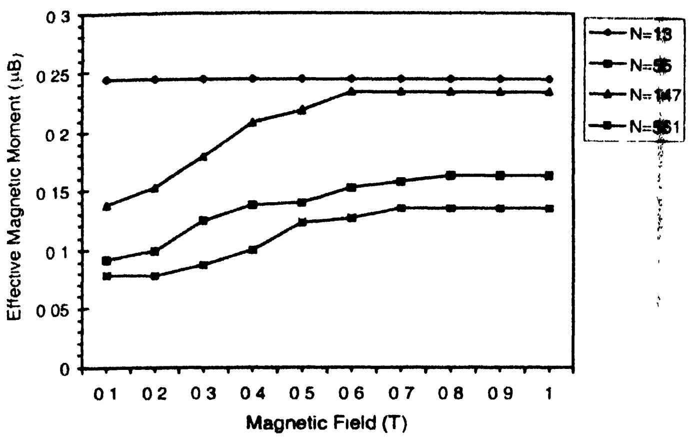

Figure 3. Size and structural dependence of the magnetic properties of cuboctahedral clusters for different applied fields with T =0 2 K

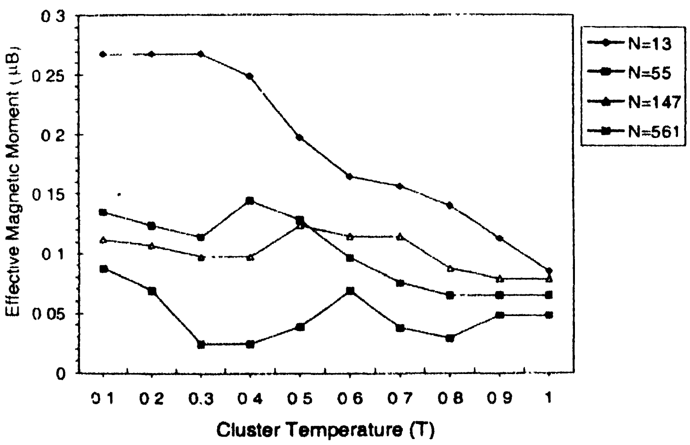

Figure 4. Size and structural dependence of the magnetic properties of cuboctahedral clusters for different temperatures with H =0.2 K.

The effective magnetic moments of small antiferromagnetic clusters have been investigated by assuming that the clusters undergo the superparamagnetic model. μ_{eff} gets saturated with the increasing applied field for icosahedral clusters but remains unchanged with applied field, for cuboctahedral clusters. The characteristic features observed are that μ_{eff}'s are larger in icosahedral and smaller in cuboctahedral clusters. This shows the structural arrangements of the clusters. It has been investigated in both icosahedral and cuboctahedral clusters that μ_{eff} increases linearly with cluster size before saturating at large size.

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